## Polaron Formation and Colossal Magnetoresistance in Manganites

he recent discovery of huge magnetoresistance effects in the manganese oxide class of materials (such as  $La_{l-x}A_xMnO_3$  (A = Sr, Ca, Ba)) has rekindled intense interest in these systems for three reasons. One is that this "colossal magnetoresistance" (CMR) offers potential in a number of technologies, such as for read/write heads in magnetic recording media, sensors, and spin-polarized electronics. The second reason is that these materials are interesting from a fundamental viewpoint. In contrast to traditional isotropic ferromagnets such as Fe, Co, Ni, and EuO where the spin system is isolated from the lattice, in the manganites the charge, spin, and lattice degrees of freedom are strongly coupled together, leading to a delicate balance of interactions that gives rise to a rich variety of physical phenomena of current interest in condensed matter science. These include a metal-insulator transition that is juxtaposed with ferromagnetism, charge ordering, orbital ordering, polaron formation, electronic phase separation, and spin and charge stripes. The third reason is because of the similarity to the high T cuprate oxides. Recent progress in our understanding of the cuprates has provided insights into the manganites, and a deeper understanding of the fundamental properties of the manganites will surely elucidate the shared concepts underlying both classes of materials.

The electrical conductivity in these manganites is directly connected to the magnetic system through the double exchange mechanism, while the Jahn-Teller distortion of the Mn³+O<sub>6</sub> couples the magnetic and lattice systems, resulting in the electronic, lattice, and magnetic degrees of freedom being intimately intertwined. The ferromagnetic transition then is non-conventional, and typically coincides with a metal-insulator transition to produce CMR. The present measurements on La<sub>0.7</sub>Ca<sub>0.3</sub>MnO<sub>3</sub>, which is near optimal doping for CMR, show that polarons form as the ferro- to paramagnetic transition is traversed, self-trapping the carriers. These polarons then "melt" when a magnetic field is applied, freeing the carriers and generating the CMR.

The scattering from a lattice polaron arises from the structural distortion that surrounds a carrier and traps it. Individual polarons generate diffuse scattering around the fundamental Bragg peaks, while polaron correlations will give rise to additional superlattice peaks. We observe both of these effects [1], and in Fig. 1 we show the four satellite



FIGURE 1. Polaron correlation peaks near the 5 0 0 Bragg peak at 270 K, just above  $T_{\rm c}$  = 255 K.

peaks at  ${}^{1}\!\!/_{4}$   ${}^{1}\!\!/_{4}$  0 positions near the 5 0 0 Bragg peak, just above  $T_{\rm c}$  (255 K). The intrinsic widths of these peaks indicate that the correlations are short range ( $\approx 15$  Å) in nature, and this range is found to be only weakly temperature dependent. Polarized neutron measurements show that these peaks originate from lattice distortions rather than magnetic scattering. Similar direct evidence for polaron formation and correlations has been observed in the bilayer manganite  ${\rm La_{1.2}Sr_{1.8}Mn_2O_7}$  [2], although the ordering wave vectors and the ratios of diffuse scattering to correlated scattering are different.

A model that can explain both the ordering wave vector as well as the overall behavior of the observed intensities of the short-range correlation peaks is the CE model developed for the half-doped case. The CE model has an orbitally-ordered Jahn-Teller lattice with charge stripes in the [110] direction, and an antiferromagnetic ground state. Such ordering has been observed for  $x \ge 0.5$  at low temperatures. This behavior contrasts with the present ferromagnetic system, where the orbital/charge correlations are short range in nature, occur only above  $T_{\rm c}$ , and are not accompanied by any antiferromagnetic correlations. The CE "stripes" are also consistent with stripe formation as seen in other transition metal oxides (nickelates and cuprates).

Of particular interest is how the polaron ordering is related to the metal-insulator transition and the quasielastic

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"central peak" [3] associated with the spin component of the polaron. As we see in Fig. 2, the temperature dependence of the intensity of the charge ordering is nearly identical to the quasielastic magnetic peak, and both follow the resistivity behavior. This is a very strong indication that these properties have a common origin in polaron formation.

A further indication of the strong relationship between charge ordering and resistivity is established by measurements in magnetic fields [1]. In Fig. 3 we see that the intensity of the charge ordering peaks decreases rapidly with increasing field. Thus the polarons "melt" when the magnetization develops and the conductivity increases, either by lowering the temperature (Fig. 2) or increasing the applied field, producing CMR. Identical field dependence has been observed both for the central peak in La<sub>0.7</sub>Ca<sub>0.3</sub>MnO<sub>3</sub> [1,3] as well as the polaron peaks in the bilayer system [2].

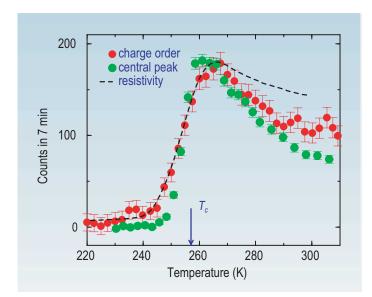


FIGURE 2. Temperature dependence of the intensity of the polaron peak at 3.75 0.25 0, compared to the magnetic quasielastic central peak, and the resistivity. The data have been scaled vertically so the peak heights match. The similarity of the data indicates a common physical origin.

The above experimental results reveal that both the spin and charge correlations associated with the polarons in La<sub>0.7</sub>Ca<sub>0.3</sub>MnO<sub>3</sub> appear together, and have a very similar temperature dependence. This coincidence may explain the amplified magnetoresistive effects, as well as the absence of conventional magnetic critical behavior. The physical

quantities that control the characteristic temperature for polaron formation, including the role that intrinsic inhomogeneities, stress, and phase separation play in their formation, still remain to be elucidated.

## **References**

- C. P. Adams, J. W. Lynn, Y. M. Mukovskii, A. A. Arsenov, and D. A. Shulyatev, Phys. Rev. Lett. 85, 3954 (2000); J. Appl. Phys. 89, 6846 (2001).
- [2] L. Vasiliu-Doloc, S. Rosenkranz, R. Osborn, S. K. Sinha, J. W. Lynn, J. Mesot, O. Seeck, G. Preosti, A. J. Fedro, and J. F. Mitchell, Phys. Rev. Lett. 83, 4393 (1999).
- [3] J. W. Lynn, R. W. Erwin, J. A. Borchers, Q. Huang, A. Santoro, J. L. Peng, and Z. Y. Li, Phys. Rev. Lett. 76, 4046 (1996); J. Appl. Phys. 81, 5488 (1997).

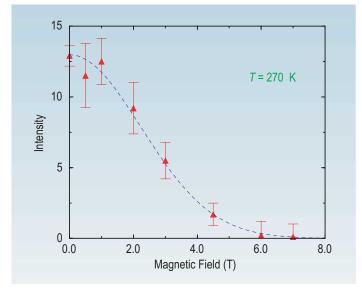


FIGURE 3. Magnetic field dependence of the intensity of the polaron peak, showing that the polarons melt with increasing field.